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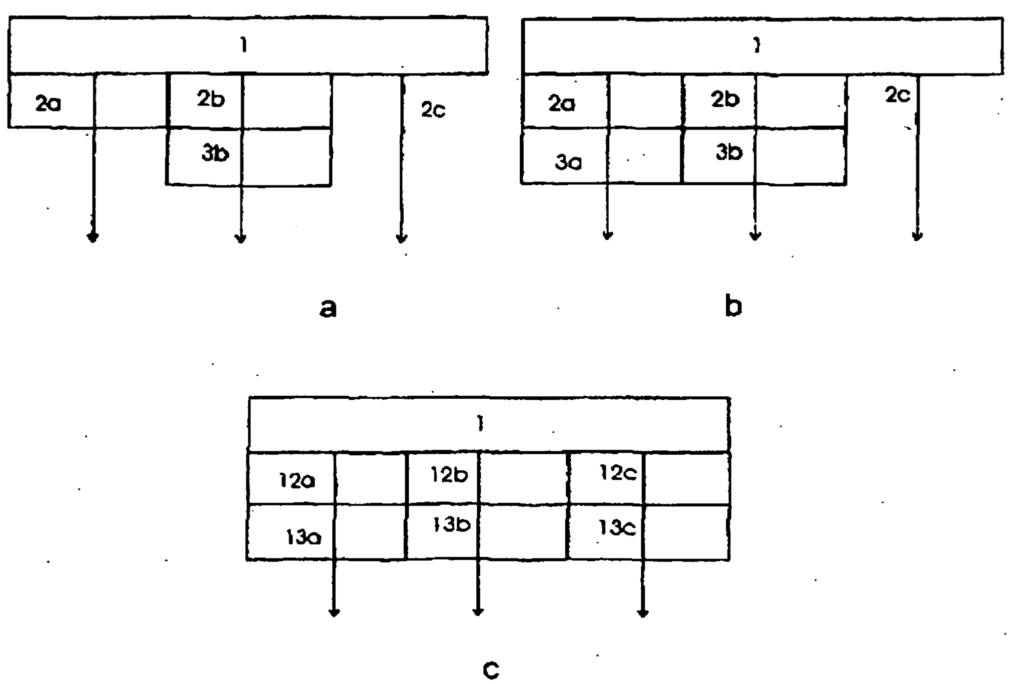
(54) Title: ELECTROLUMINESCENT DEVICES



(57) Abstract: An electroluminescent device has a layer which emits light in the blue, purple/blue or ultraviolet section of the spectrum and a layer which contains a fluorescent material and optionally a layer comprising one or more colour filters so that light emitted by the electroluminescent layer excites the fluorescent material causing light to be emitted at a longer wavelength.

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Electroluminescent Devices

The present invention relates to an electroluminescent device which can emit light of different colours.

Materials which emit light when an electric current is passed through them are well known and used in a wide range of display applications. Liquid crystal devices and devices which are based on inorganic semiconductor systems are widely used, however these suffer from the disadvantages of high energy consumption, high cost of manufacture, low quantum efficiency and the inability to make flat panel displays.

Typical electroluminescent devices comprise an anode, normally of an electrically light transmitting material, a layer of a hole transporting material, a layer of the electroluminescent material, optionally a layer of an electron transmitting material and a metal cathode. There can be other layers, such as buffer layers and the layers can be combined using mixtures of one or more of the hole transporting material, electroluminescent material and the electron transmitting material.

Patent application WO98/58037 describes a range of lanthanide complexes which can be used in electroluminescent devices which have improved properties and give better results. Patent Applications PCT/GB98/01773, PCT/GB99/03619, PCT/GB99/04030, PCT/GB99/04028, PCT/GB00/00268 describe electroluminescent complexes, structures and devices using rare earth chelates.

For some applications light in several colours is required to be emitted e.g. red, green and blue of specified wavelengths. One way of accomplishing this is to have three electroluminescent structures side by side each of electroluminescent structures emits light of the appropriate wavelength, however such devices are difficult to make and operate.

We have now devised a device which can emit light at several wavelengths which uses one electroluminescent structure.

According to the invention there is provided an electroluminescent device which comprises (i) an electroluminescent structure which emits light in the blue, purple/blue or ultraviolet section of the spectrum (ii) a fluorescent layer comprising one or more fluorescent sections side by side each of which sections incorporates a fluorescent material and optionally (iii) a layer comprising one or more colour filters in which device light emitted by the electroluminescent structure excites a fluorescent material in the fluorescent layer causing light to be emitted at a longer wavelength.

For example the fluorescent layer can comprise a first section which contains a fluorescent material which will fluoresce in the red region of the spectrum when excited by ultra-violet or blue light, a second section which contains a fluorescent material which will fluoresce in the green region of the spectrum and a third section which contains a fluorescent material which will fluoresce in the blue region of the spectrum when the incident light is in the ultra-violet but which need not incorporate any fluorescent material if the blue light emitted by the electroluminescent structure is of the appropriate wavelength.

If, due to the nature of the fluorescent materials, the wavelengths emitted by these materials needs further modification there can be a colour filter through which the light passes.

When the emitted light is in the blue spectrum preferably the peak wavelength of the light is 430 to 440 nm e.g. 440 nm and when the emitted light is in the lighter blue region of the spectrum the emitted light has a peak wavelength of about 470 nm.

The colour of light is subjective and colours can be defined by co-ordinates on a two dimensional chart in which colours are areas on the chart and in the present invention

the blue and purple blue spectrum is defined as the area bounded by the co-ordinates in the colour chart CIE I931. Suitable complexes emit light within the co-ordinates (0, 0) (0, 0.3) (0.3, 0).

Compounds which emit light in the blue or purple blue region of the spectrum include lithium quinolate.

Patent Application WO 00/32717 discloses the use of lithium quinolate as an electroluminescent material in electroluminescent devices. Lithium quinolate emits light in the blue spectrum and has greater electron mobility, of the order of 45%, than the widely used aluminium quinolate and aluminium quinolate derivatives which can make it a more effective electroluminescent material.

As described in this patent application the lithium quinolate is preferably synthesised by the reaction, in an inert solvent, e.g. acetonitrile, of 8-hydroxyquinoline with a lithium alkyl e.g. n-butyl lithium. The lithium quinolate is an off white or white solid at room temperature.

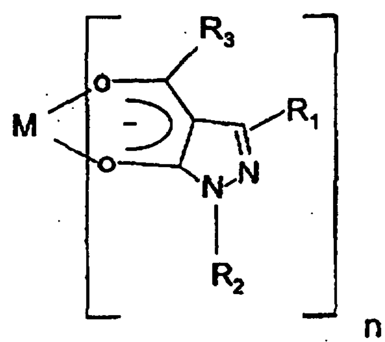
An article by C. Schmitz, H Scmidt and M. Thekalakat entitled Lithium Quinolate Complexes as Emitter and Interface Materials in Organic Light-Emitting Diodes in Chem. Mater, 2000, 12, 3012-3019 discloses the use of a layer of lithium quinolate together with hole transporting materials in electroluminescent devices.

As well as the lithium salt of 8-hydroxyquinoline, the term quinolate in this specification includes salts of substituted 8-hydroxyquinoline.

The blue colour emitted by lithium quinolate can be made a deeper blue if there is a layer of lithium quinolate mixed with the hole transporting compound α-NPB.

Other compounds which emit light in the blue region of the spectrum are non rare earth metal chelates such as the metal tris-(1,3-diphenyl-1-l-propanedione) (DBM) complexes where the metal can include aluminium, scandium, magnesium, zinc etc. For example $Al(DBM)_3$, $Zn(DBM)_2$ and $Mg(DBM)_2$, $Sc(DBM)_3$ etc. Such complexes are described in patent Application PCT/GB02/02722.

Other metal complexes which can emit light in the blue region of the spectrum are



where M is a metal; n is the valency of M; R_1, R_2 and R_3 which may be the same or different are selected from hydrogen, hydrocarbyl groups, substituted and unsubstituted aliphatic groups substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorocarbons such as trifluoryl methyl groups, halogens such as fluorine or thiophenyl groups or nitrile; R_1, and R_3 can also be form ring structures and R_1, R_2 and R_3 can be copolymerisable with a monomer e.g. styrene.

Preferably M is aluminium, beryllium or boron.

Such complexes are described in Patent Application PCT/GB02/0032163.

Other complexes are described in WO 00/26321, these complexes are TMHD (Tris(2,2,6,6-tetramethyl-3,5-heptanedionato), tripyridyl, bathophen (4,7-diphenyl-

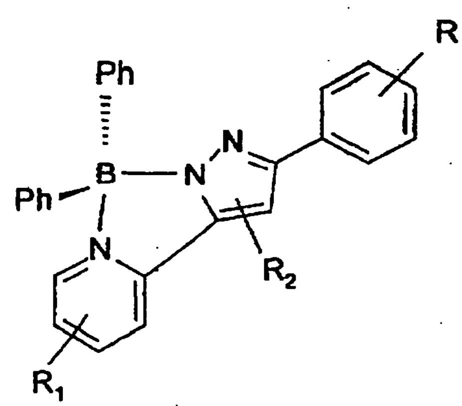
1,1-phenanthroline) complexes of metals such as thorium (IV), yttrium (III), gadolinium (III), europium (II), terbium(IV), cerium(IV) and cerium (III). A mixture of metals can be used to form mixed chelates.

Preferred complexes are Eu(II)$(TMHD)_2$.

Compounds which emit light in the ultra-violet region of the spectrum are described in WO 00/44851 and preferably are organic complexes of gadolinium in the III state. The preferred complexes are complexes with ligands such as ethylene diamine tetramine EDTA, DCTA, DTPA and TTHA the formulae of which are shown in figure 1 of the drawings.

Particularly preferred mixed complexes are the gadolinium complexes e.g. Gd$[Eu(EDTA)]_3$ and Na$[Gd(EDTA)_3]$.

Other electroluminescent complexes which emit light in the blue region of the spectrum include boron complexes of formula



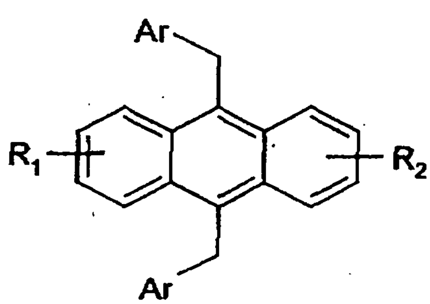
where Ph is an unsubstituted or substituted phenyl group where the substituents can be the same or different and are selected from hydrogen, and substituted and

unsubstituted hydrocarbyl groups such as substituted and unsubstituted aliphatic groups, substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorocarbons such as trifluoryl methyl groups, halogens such as fluorine or thiophenyl groups; R, R_1 and R_2 can be hydrogen or substituted or unsubstituted hydrocarbyl groups, such as substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorine, fluorocarbons such as trifluoryl methyl groups, halogens such as fluorine or thiophenyl groups or nitrile.

Examples of R and/or R_1 and/or R_2 and/or R_3 include aliphatic, aromatic and heterocyclic alkoxy, aryloxy and carboxy groups, substituted and substituted phenyl, fluorophenyl, biphenyl, phenanthrene, anthracene, naphthyl and fluorene groups alkyl groups such as t-butyl, heterocyclic groups such as carbazole.

These complexes are described in GB Patent Application GB0306097.7.

Other blue emitters are anthracene compounds of formula



(A)

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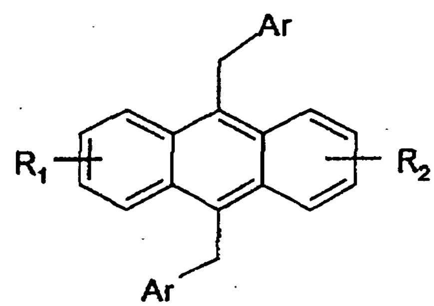

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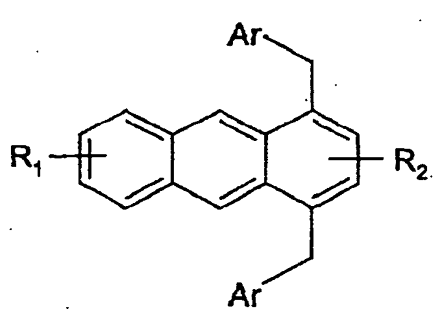

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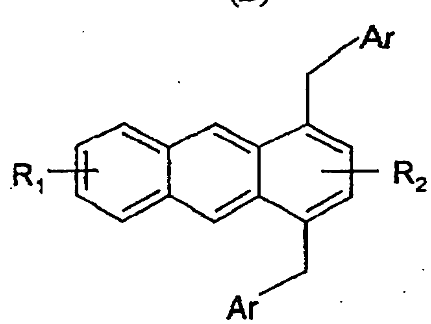


(D)

where Ar is an aromatic or a substituted aromatic group or a tertiary alkyl group such as t-butyl and R_1 and R_2 are the same or different and are selected from hydrogen, and substituted and unsubstituted hydrocarbyl groups such as substituted and unsubstituted aliphatic groups, substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorocarbons such as trifluoryl methyl groups, halogens such as fluorine or thiophenyl groups; R_1, and R_2 can also form substituted and unsubstituted fused aromatic, heterocyclic and polycyclic ring structures and can be copolymerisable with a monomer e.g. styrene.

These complexes are described in GB Patent Application 0227376.1.

Compounds which fluoresce in the red and green region of the spectrum when excited by blue or ultra-violet light are well known for red light; one class includes europium complexes and for green light one class includes terbium complexes.

The electroluminescent structure preferably comprises sequentially a first electrode which functions as the cathode, a layer of an electroluminescent material which emits light in the blue, purple/blue or ultraviolet section of the spectrum and a second electrode which functions as the anode.

Preferably the anode is transparent and light is emitted through the anode and layer (ii) is adjacent the anode.

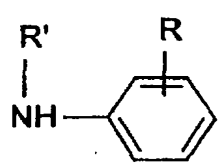
Preferably there is a layer of an electron injecting material between the cathode and the electroluminescent material layer; the electron injecting material is a material which will transport electrons when an electric current is passed through electron injecting materials include a metal complex such as a metal quinolate e.g. an aluminium quinolate, lithium quinolate, a cyano anthracene such as 9,10 dicyano anthracene, cyano substituted aromatic compounds, tetracyanoquinidodimethane a polystyrene sulphonate or a compound with the structural formulae shown in figures

3 or 4 of the drawings in which the phenyl rings can be substituted with substituents R as defined above. Instead of being a separate layer the electron injecting material can be mixed with the electroluminescent material and co-deposited with it.

Preferably there is a layer of a hole transporting material between the anode and the layer of the electroluminescent compound.

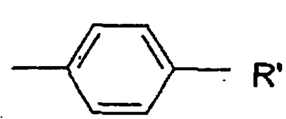
The hole transporting material can be any of the hole transporting materials used in electroluminescent devices.

The hole transporting material can be an amine complex such as poly (vinylcarbazole), N, N'-diphenyl-N, N'-bis (3-methylphenyl) -1,1' -biphenyl -4,4'-diamine (TPD), an unsubstituted or substituted polymer of an amino substituted aromatic compound, a polyaniline, substituted polyanilines, polythiophenes, substituted polythiophenes, polysilanes etc. Examples of polyanilines are polymers of



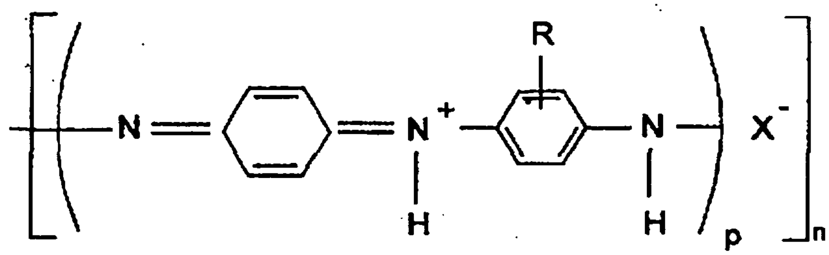
(XXVI)

where R is in the ortho – or meta-position and is hydrogen, C1-18 alkyl, C1-6 alkoxy, amino, chloro, bromo, hydroxy or the group



where R is alky or aryl and R' is hydrogen, C1-6 alkyl or aryl with at least one other monomer of formula I above.

Or the hole transporting material can be a polyaniline, polyanilines which can be used in the present invention have the general formula



(XXVII)

where p is from 1 to 10 and n is from 1 to 20, R is as defined above and X is an anion, preferably selected from Cl, Br, SO_4, BF_4, PF_6, H_2PO_3, H_2PO_4, arylsulphonate, arenedicarboxylate, polystyrenesulphonate, polyacrylate alkysulphonate, vinylsulphonate, vinylbenzene sulphonate, cellulose sulphonate, camphor sulphonates, cellulose sulphate or a perfluorinated polyanion.

Examples of arylsulphonates are p-toluenesulphonate, benzenesulphonate, 9,10-anthraquinone-sulphonate and anthracenesulphonate, an example of an arenedicarboxylate is phthalate and an example of arehecarboxylate is benzoate.

We have found that protonated polymers of the unsubstituted or substituted polymer of an amino substituted aromatic compound such as a polyaniline are difficult to evaporate or cannot be evaporated. However we have surprisingly found that if the unsubstituted or substituted polymer of an amino substituted aromatic compound is deprotonated then it can be easily evaporated i.e. the polymer is evaporable.

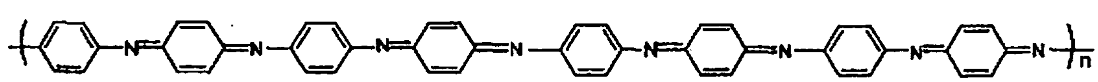
Preferably evaporable deprotonated polymers of unsubstituted or substituted polymer of an amino substituted aromatic compound are used. The de-protonated unsubstituted or substituted polymer of an amino substituted aromatic compound can be formed by deprotonating the polymer by treatment with an alkali such as ammonium hydroxide or an alkali metal hydroxide such as sodium hydroxide or potassium hydroxide.

The degree of protonation can be controlled by forming a protonated polyaniline and de-protonating. Methods of preparing polyanilines are described in the article by A. G. MacDiarmid and A. F. Epstein, Faraday Discussions, Chem Soc.88 P319 1989.

The conductivity of the polyaniline is dependant on the degree of protonation with the maximum conductivity being when the degree of protonation is between 40 and 60% e.g. about 50% for example.

Preferably the polymer is substantially fully deprotonated.

A polyaniline can be formed of octamer units i.e. p is four e.g.



The polyanilines can have conductivities of the order of 1 x 10^{-1} Siemen cm^{-1} or higher.

The aromatic rings can be unsubstituted or substituted e.g. by a C1 to 20 alkyl group such as ethyl.

The polyaniline can be a copolymer of aniline and preferred copolymers are the copolymers of aniline with o-anisidine, m-sulphanilic acid or o-aminophenol, or o-

toluidine with o-aminophenol, o-ethylaniline, o-phenylene diamine or with amino anthracenes.

Other polymers of an amino substituted aromatic compound which can be used include substituted or unsubstituted polyaminonapthalenes, polyaminoanthracenes, polyaminophenanthrenes, etc. and polymers of any other condensed polyaromatic compound. Polyaminoanthracenes and methods of making them are disclosed in US Patent 6,153,726. The aromatic rings can be unsubstituted or substituted e.g. by a group R as defined above.

Other hole transporting materials are conjugated polymer and the conjugated polymers which can be used can be any of the conjugated polymers disclosed or referred to in US 5807627, PCT/WO90/13148 and PCT/WO92/03490.

The preferred conjugated polymers are poly (p-phenylenevinylene)-PPV and copolymers including PPV. Other preferred polymers are poly(2,5 dialkoxyphenylene vinylene) such as poly (2-methoxy-5-(2-methoxypentyloxy-1,4-phenylene vinylene), poly(2-methoxypentyloxy)-1,4-phenylenevinylene), poly(2-methoxy-5-(2-dodecyloxy-1,4-phenylenevinylene) and other poly(2,5 dialkoxyphenylenevinylenes) with at least one of the alkoxy groups being a long chain solubilising alkoxy group, poly fluorenes and oligofluorenes, polyphenylenes and oligophenylenes, polyanthracenes and oligo anthracenes, ploythiophenes and oligothiophenes.

In PPV the phenylene ring may optionally carry one or more substituents e.g. each independently selected from alkyl, preferably methyl, alkoxy, preferably methoxy or ethoxy.

Any poly(arylenevinylene) including substituted derivatives thereof can be used and the phenylene ring in poly(p-phenylenevinylene) may be replaced by a fused ring

system such as anthracene or naphthlyene ring and the number of vinylene groups in each polyphenylenevinylene moiety can be increased e.g. up to 7 or higher.

The conjugated polymers can be made by the methods disclosed in US 5807627, PCT/WO90/13148 and PCT/WO92/03490.

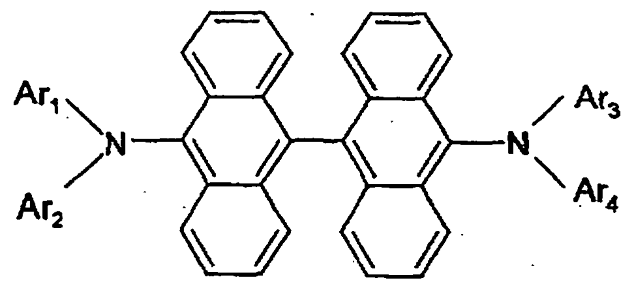
The thickness of the hole transporting layer is preferably 20nm to 200nm.

The polymers of an amino substituted aromatic compound such as polyanilines referred to above can also be used as buffer layers with or in conjunction with other hole transporting materials.

The structural formulae of some other hole transporting materials are shown in Figures 2, and 5 - 8 of the drawings, where R_1, R_2 and R_3 can be the same or different and are selected from hydrogen, and substituted and unsubstituted hydrocarbyl groups such as substituted and unsubstituted aliphatic groups, substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorocarbons such as trifluoryl methyl groups, halogens such as fluorine or thiophenyl groups; R_1, R_2 and R_3 can also form substituted and unsubstituted fused aromatic, heterocyclic and polycyclic ring structures and can be copolymerisable with a monomer e.g. styrene. X is Se, S or O, Y can be hydrogen, substituted or unsubstituted hydrocarbyl groups, such as substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorine, fluorocarbons such as trifluoryl methyl groups, halogens such as fluorine or thiophenyl groups or nitrile.

Examples of R_1 and/or R_2 and/or R_3 include aliphatic, aromatic and heterocyclic alkoxy, aryloxy and carboxy groups, substituted and substituted phenyl, fluorophenyl, biphenyl, phenanthrene, anthracene, naphthyl and fluorene groups alkyl groups such as t-butyl, heterocyclic groups such as carbazole.

Other hole transporting materials are diamino dianthracenes of formula



where Ar_1, Ar_2, Ar_3 and Ar_4 are the same or different substituted or unsubstituted aromatic groups including substituted or unsubstituted monocyclic or polycyclic aromatic groups such as phenyl, naphthyl, phenanthrenyl etc.

The preferred groups Ar_1, Ar_2, Ar_3 and Ar_4 are substituted and unsubstituted phenyl, bisphenyl, naphthyl, anthracenyl, and fused rings where Ar_1 and Ar_2 or Ar_3 and Ar_4 form heterocyclic ring with the nitrogen atom. The substituents can be selected from hydrogen, and alkyl, aliphatic, aromatic and heterocyclic alkoxy, aryloxy and carboxy groups, such as t-butyl and heterocyclic groups such as carbazole and trimethyl fluorine.

Optionally the hole transporting material can be mixed with the electroluminescent material and co-deposited with it.

The hole transporting materials, the electroluminescent material and the electron injecting materials can be mixed together to form one layer, which simplifies the construction.

The anode is preferably a transparent substrate such as a conductive glass or plastic material which acts as the anode. Preferred substrates are conductive glasses such as indium tin oxide coated glass, but any glass which is conductive or has a conductive layer such as a metal or conductive polymer can be used. Conductive polymers and conductive polymer coated glass or plastics materials can also be used as the

substrate.

The cathode is preferably a low work function metal e.g. aluminium, calcium, lithium, silver/magnesium alloys, rare earth metal alloys, magnesium/aluminium, magnesium/ indium, titanium/aluminium, etc., aluminium is a preferred metal. A metal fluoride such as an alkali metal, rare earth metal or their alloys can be used as the second electrode for example by having a metal fluoride layer formed on a metal.

An electroluminescent structure is shown in fig. 9 in which (1) is an aluminium cathode, (2) is an electron transmitting layer, (3) is a layer of an electroluminescent compound which emits light in the blue, purple blue or ultra-violet region of the spectrum, (4) is a layer of a hole transporting material and (5) is an indium tin oxide coated glass anode.

Devices according to the invention are described in figs. 10a to 10c of the drawings in which (1) is an electroluminescent structure shown in fig. 9; in fig. 10a (1) emits light in the blue region of the spectrum at about 470nm (2a) is a layer containing a red fluorescent, (2b) is a green fluorescent and (3b) is a green filter.

The electroluminescent structure is turned on so that it emits blue light of the required wavelength, the light is incident on (2a), which causes it to emit red light, and on (2b) so that it emits green light. The red light emitted has the required wavelength so no filter is needed, but the filter (3c) ensures that the green light has the required wavelength. The light is emitted at the three required wavelengths to give red, green and blue light.

In fig. 10b the red fluorescent light is not at the required wavelength so a red filter (3a) is added.

In fig. 10c the electroluminescent structure (1) emits light in the ultra-violet region of the spectrum by having a layer of $Na[Gd(EDTA)_3]$ as layer (3) in fig. 9.

Layer (12a) incorporates a compound which fluoresces red when excited by ultra-violet light, (12b) incorporates a compound which fluoresces green when excited by ultra-violet light and (12c) incorporates a compound which fluoresces in the blue spectrum when excited by ultra-violet light. Filters (13a), (13b) and (13c) which are red, green and blue filters respectively can adjust the wavelength of the emitted if required.

Claims

1. An electroluminescent device which comprises (i) an electroluminescent structure which emits light in the blue, purple/blue or ultraviolet section of the spectrum and (ii) a fluorescent layer comprising one or more fluorescent sections each of which sections incorporates a fluorescent material in which device light emitted by the electroluminescent structure excites a fluorescent material in the fluorescent layer causing light to be emitted by the fluorescent material.

2. An electroluminescent device as claimed in claim 1 in which there are a plurality of fluorescent layers side by side.

3. An electroluminescent device as claimed in claim 1 or 2 in which there is a layer (iii) which is a layer comprising one or more colour filters.

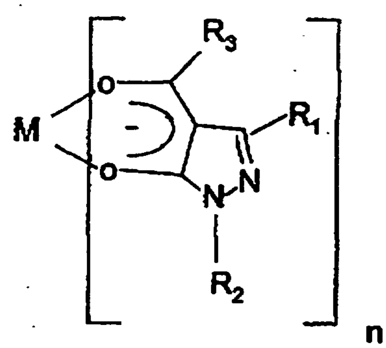
4. An electroluminescent device as claimed in any one of claims 1 to 3 in which the electroluminescent structure comprises sequentially a first electrode which functions as the cathode, a layer of an electroluminescent material which emits light in the blue, purple/blue or ultraviolet section of the spectrum and a second electrode which functions as the anode.

5. An electroluminescent device as claimed in claim 4 in which the electroluminescent material is lithium quinolate.

6. An electroluminescent device as claimed in claim 4 in which the lithium quinolate is mixed with the hole transporting compound α-NPB.

7. An electroluminescent device as claimed in claim 4 in which the electroluminescent material is $Al(DBM)_3$, $Zn(DBM)_2$ and $Mg(DBM)_2$ or $Sc(DBM)_3$.

8. An electroluminescent device as claimed in claim 4 in which the electroluminescent material is



where M is a metal; n is the valency of M; R_1, R_2 and R_3 which may be the same or different are selected from hydrogen, hydrocarbyl groups, substituted and unsubstituted aliphatic groups substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorocarbons such as trifluoryl methyl groups, halogens such as fluorine or thiophenyl groups or nitrile; R_1, and R_3 can also be form ring structures and R_1, R_2 and R_3 can be copolymerisable with a monomer e.g. styrene.

9. An electroluminescent device as claimed in claim 8 in which M is aluminium.

10. An electroluminescent device as claimed in claim 4 in which the electroluminescent material is selected from tripyridyl and bathophen (4,7-diphenyl-1,1-phenanthroline) complexes of one or more of thorium (IV), yttrium (III), gadolinium (III), europium (II), terbium(IV), cerium(IV) and cerium (III).

11. An electroluminescent device as claimed in claim 4 in which the electroluminescent material which emits light in the ultra-violet region of the spectrum is a gadolinium complex of EDTA, DCTA, DTPA and TTHA.

12. An electroluminescent device as claimed in claim 4 in which the electroluminescent material which emits light in the ultra-violet region of the spectrum is $Gd[Eu(EDTA)]_3$ or $Na[Gd(EDTA)_3]$.

13. An electroluminescent device as claimed in any one of the preceding claims in which the fluorescent compound is a europium complex which fluoresces in the red region of the spectrum when excited by blue or ultra-violet light.

14. An electroluminescent device as claimed in any one of the preceding claims in which the compound is a terbium complex which fluoresces in the green region of the spectrum when excited by blue or ultra-violet light.

15. An electroluminescent device as claimed in any one of the preceding claims in which the electroluminescent structure comprises sequentially a first electrode which functions as the cathode, a layer of a hole transporting material, a layer of an electroluminescent material which emits light in the blue, purple/blue or ultraviolet section of the spectrum, a layer of an electron transmitting material and a second electrode which functions as the anode.

16. An electroluminescent device as claimed in claim 15 in which the electron transmitting material is a metal quinolate, a cyano anthracene, 9,10 dicyano anthracene, a polystyrene sulphonate or a compound of formulae as shown in fig. 3 or 4.

17. An electroluminescent device as claimed in claim 16 in which the metal quinolate is lithium, sodium, potassium, zinc, magnesium or aluminium quinolate.

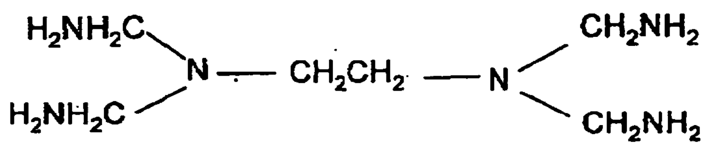
18. An electroluminescent device as claimed in claim 15 in which the hole transporting layer is an aromatic amine complex.

19. An electroluminescent device as claimed in claim 15 in which the hole transporting layer is formed from a poly(vinylcarbazole), N,N'-diphenyl-N,N'-bis (3-methylphenyl) -1,1' -biphenyl -4,4'-diamine (TPD), polyaniline, or a substituted polyaniline.

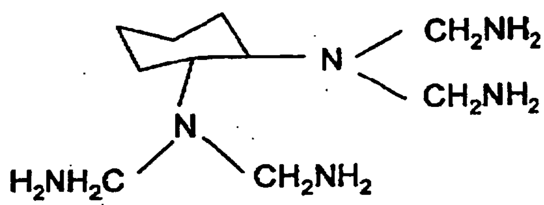
20. An electroluminescent device as claimed in claim 15 in which the hole transporting material has the formula of any of figures 1, 2 and 5 – 8.

21. An electroluminescent device as claimed in any one of the preceding claims in which the fluorescent layer comprises two fluorescent sections side by side, one of which incorporates a fluorescent material which fluoresces in the red region of the spectrum and one of which one incorporates a material which fluoresces in the green region of the spectrum and in which there is a green filter, whereby light emitted by the green fluorescent material passes through the green filter.

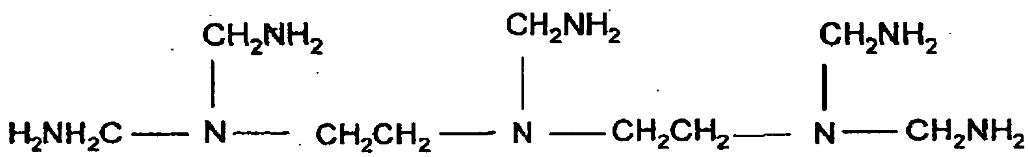
22. An electroluminescent device as claimed in any one of any one of the preceding claims in which the fluorescent layer comprises two fluorescent sections side by side, one of which incorporates a fluorescent material which fluoresces in the red region of the spectrum and one of which incorporates a material which fluoresces in the green region of the spectrum and in which there are filters whereby light emitted by the red fluorescent material passes through a red filter and light emitted by the green fluorescent material passes through a green filter.



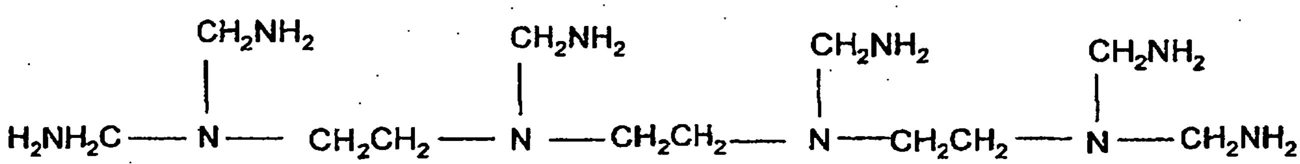
EDTA



DCTA



DTPA



TTHA

Fig. 1

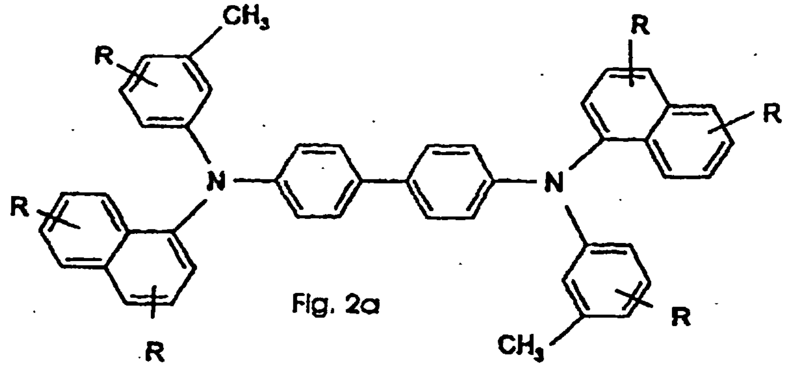


Fig. 2a

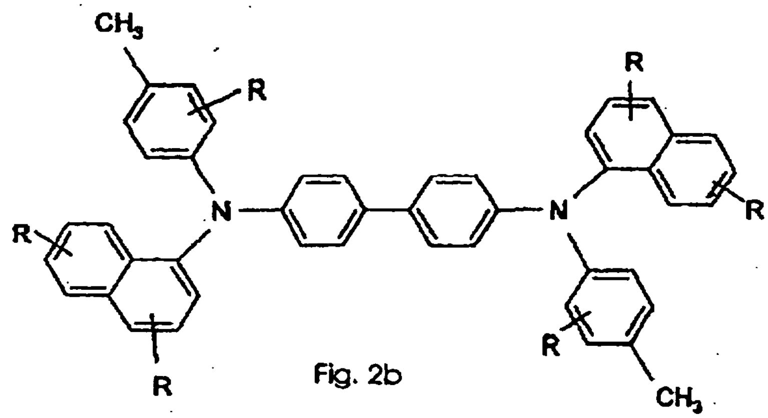


Fig. 2b

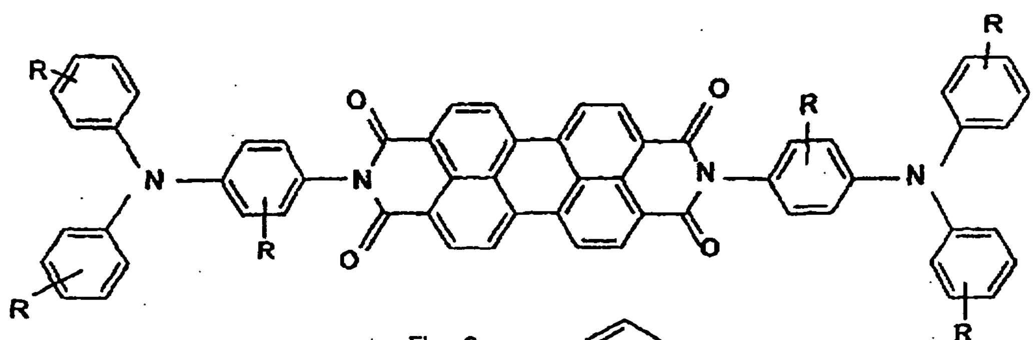


Fig. 2c

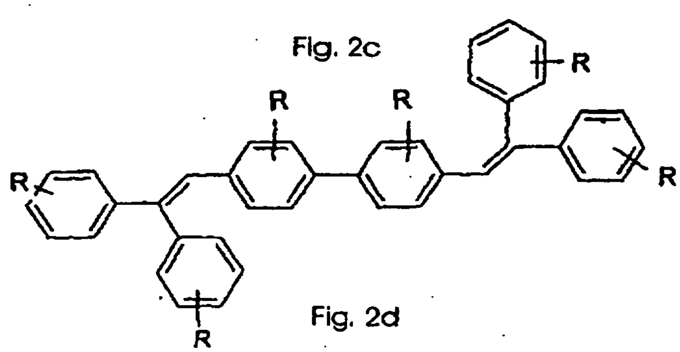
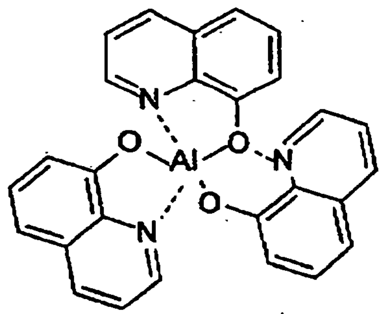
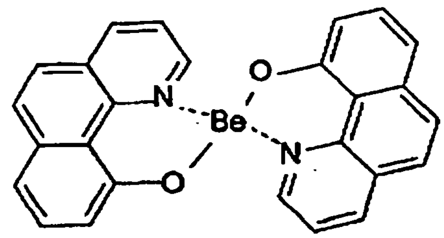


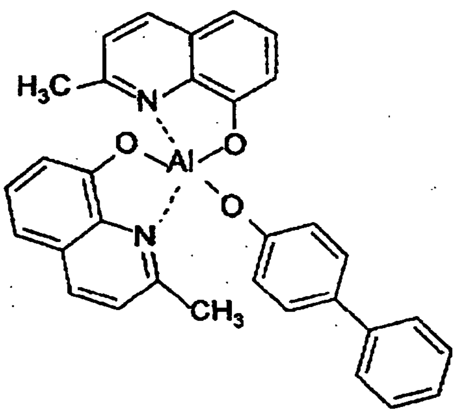
Fig. 2d



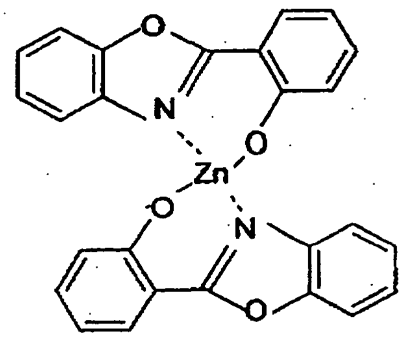
Alq



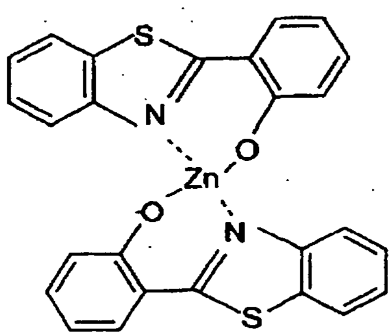
Bebq



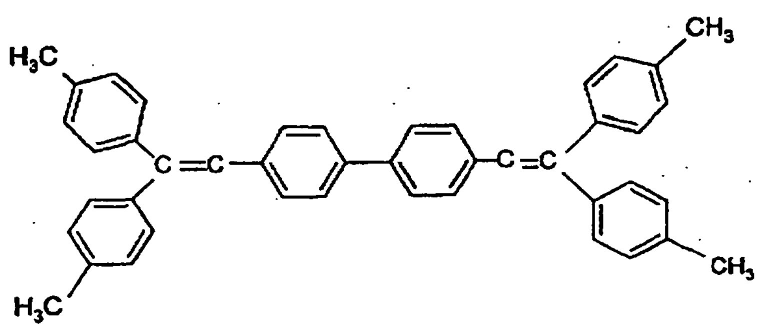
BAlq1



ZnPBO

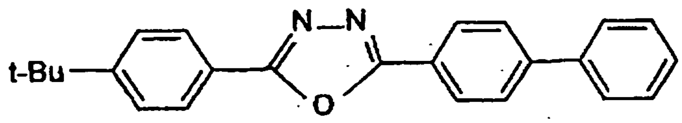


ZnPBT

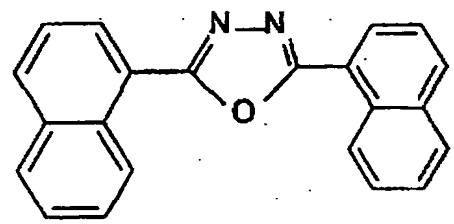


DTVb1

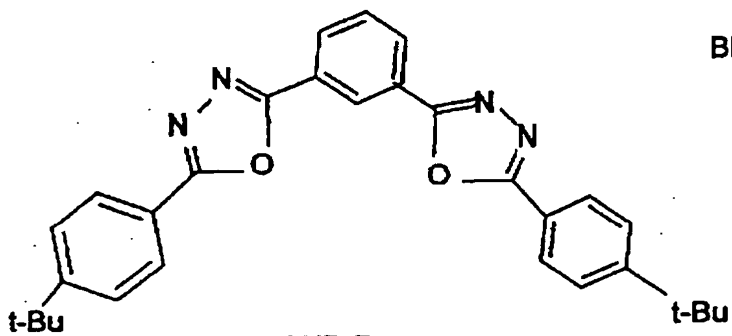
Fig. 3



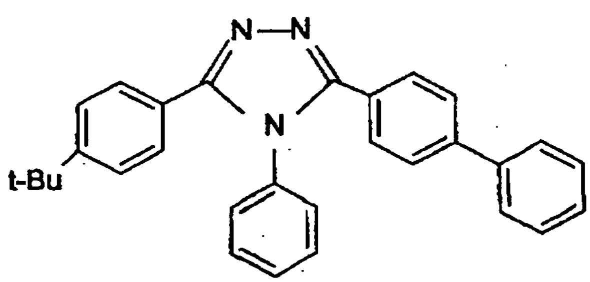
t-Bu-PBD



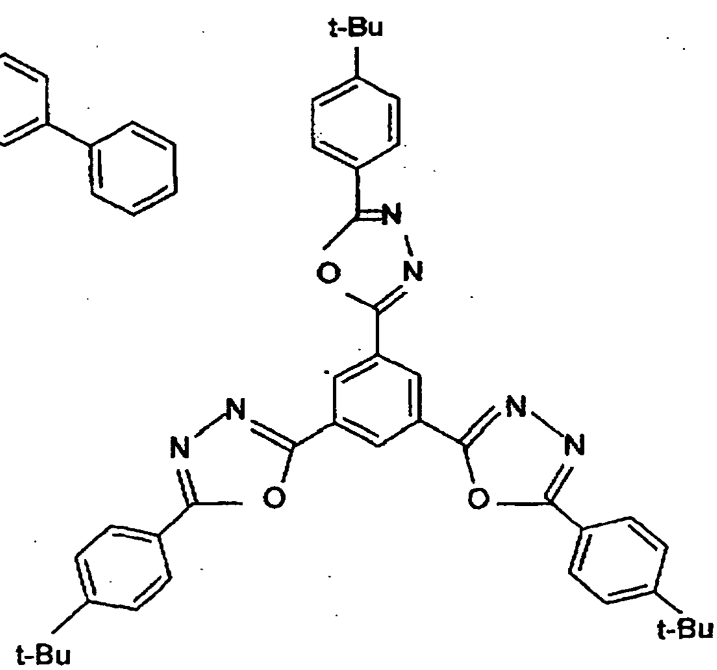
BND



OXD-7



TAZ



OXD- Star

Fig. 4

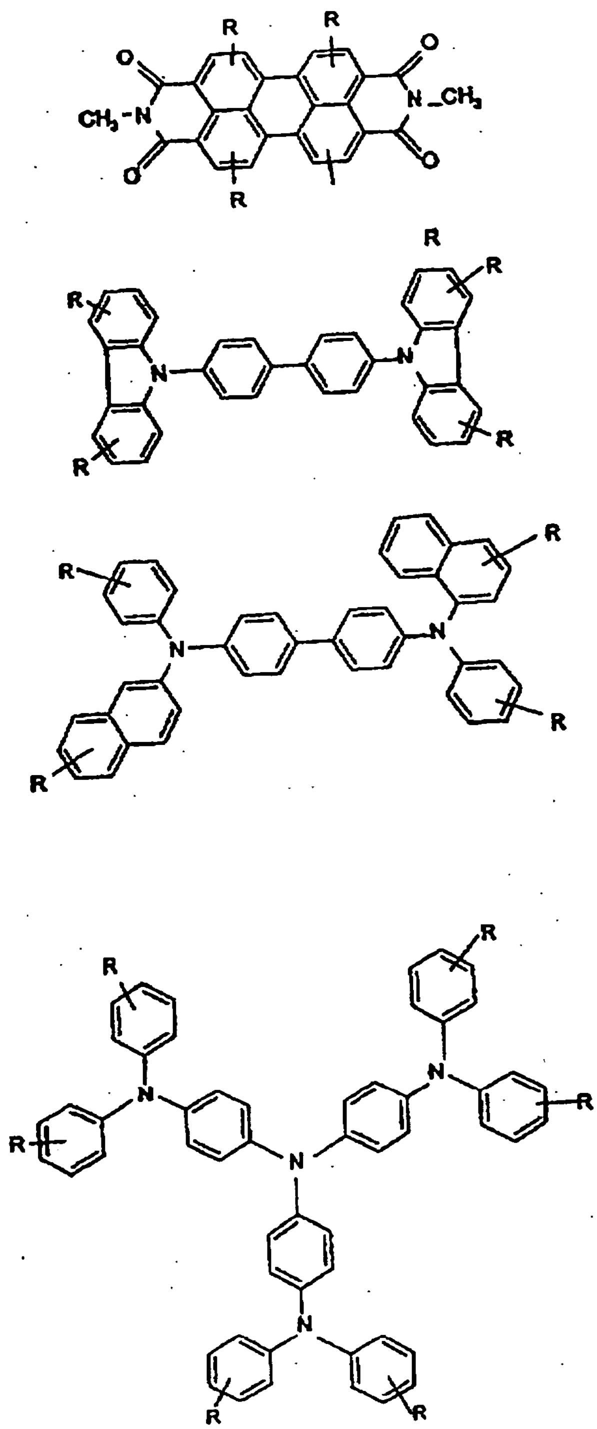
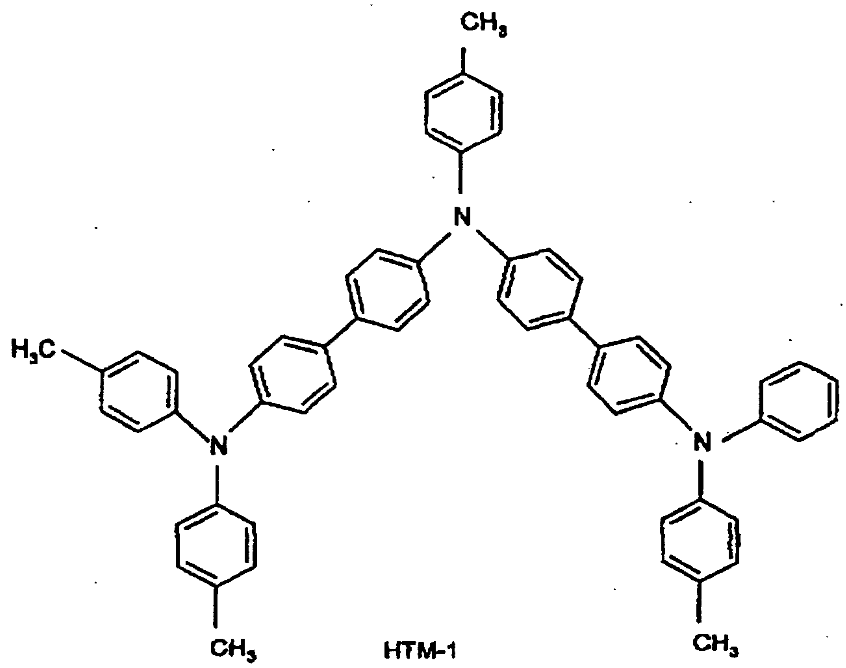
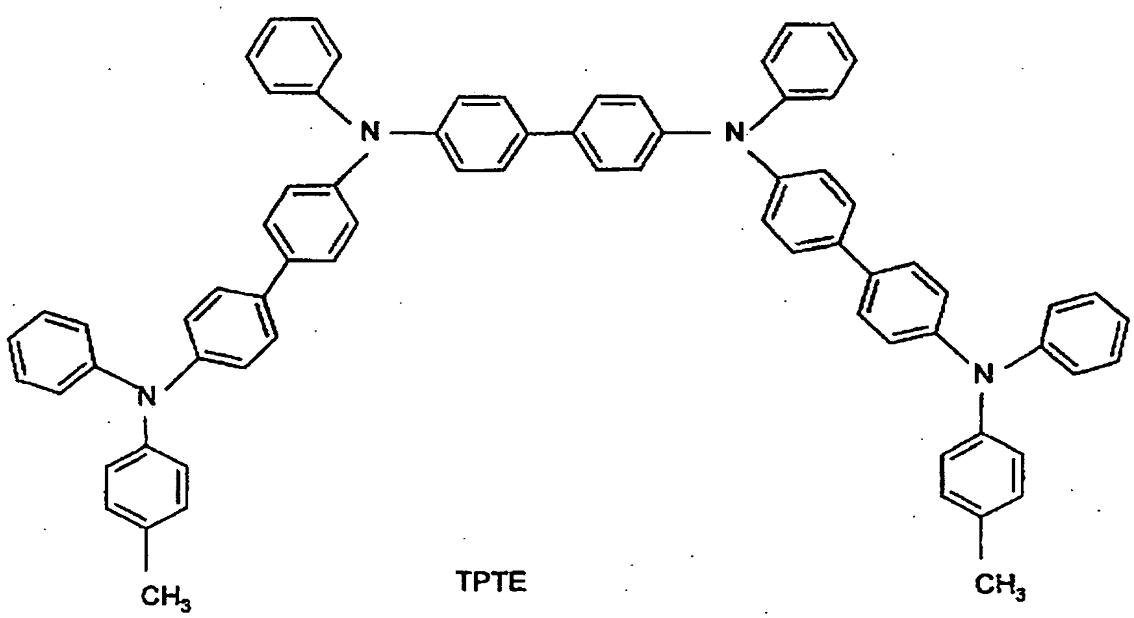


Fig. 5



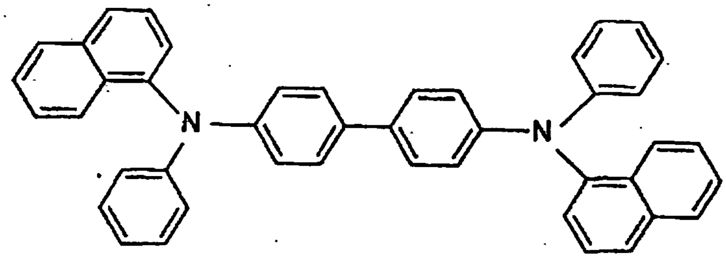
HTM-1

Fig. 15a



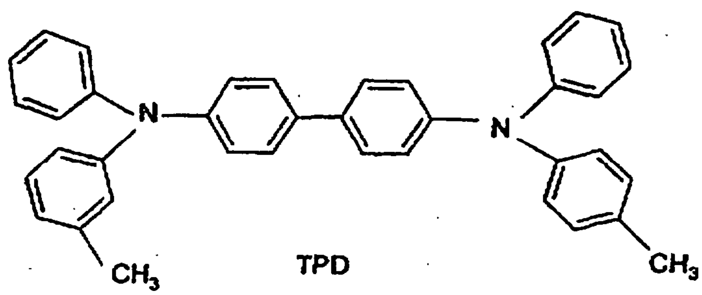
TPTE

Fig. 6



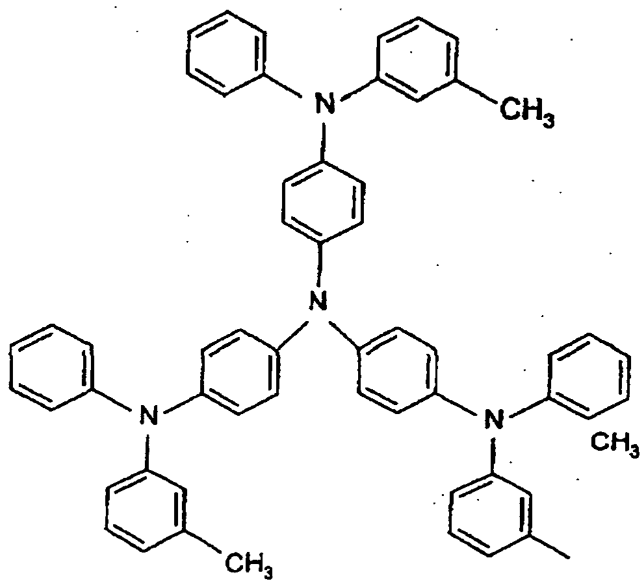
α -NPB

Fig. 7a



TPD

Fig. 7b



mTADATA

Fig. 7c

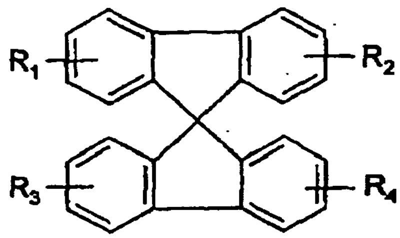


Fig. 8a

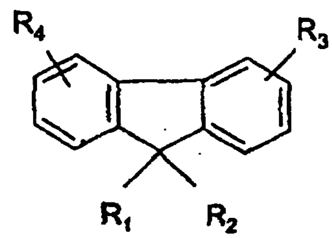


Fig. 8b

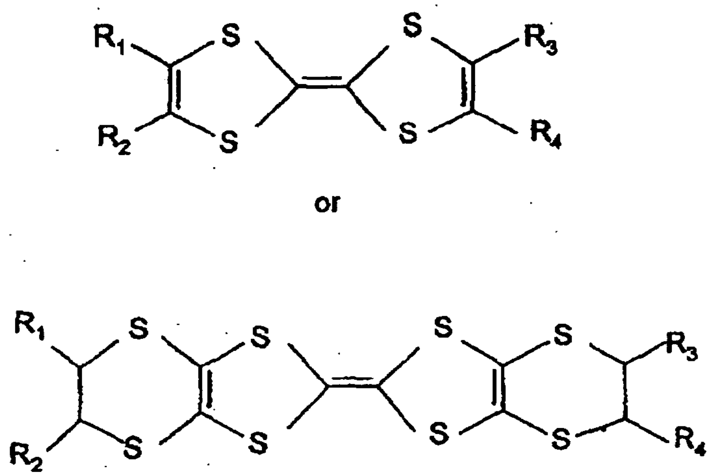


Fig. 8c

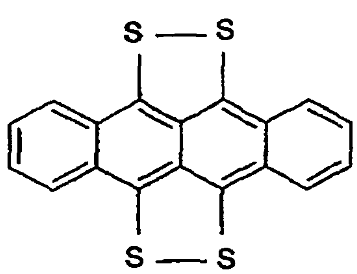


Fig. 8d

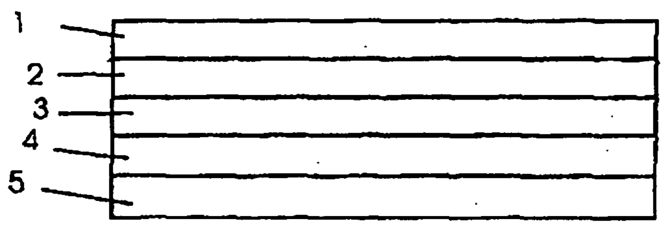


Fig. 9

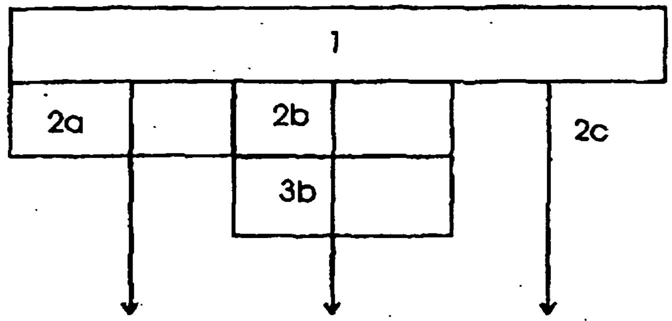


Fig. 10a

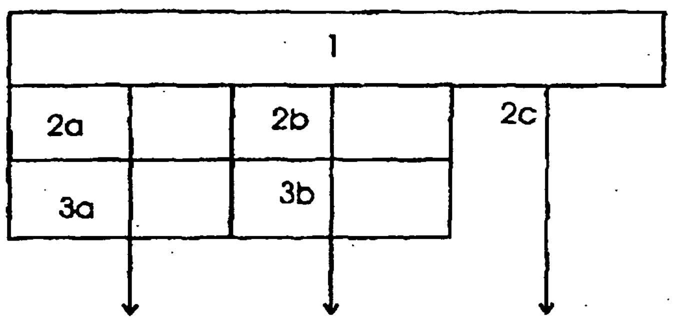


Fig. 10b

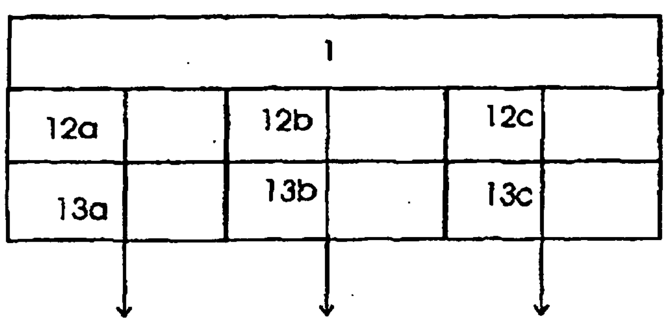


Fig. 10c

INTERNATIONAL SEARCH REPORT

International Application No
PCT/GB 03/01932

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C09K11/06 H05B33/14 H01L51/20

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 C09K H05B H01L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ, CHEM ABS Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 1 013 740 A (KONISHIROKU PHOTO IND) 28 June 2000 (2000-06-28)	1-4,10, 13,15-22
Y	claims 28-30,32,34-39; examples 3-1,3-3,5-1	5-9,11, 12,14
X	GB 2 333 897 A (FUJI ELECTRIC CO LTD) 4 August 1999 (1999-08-04) the whole document	1-4,6, 15-19, 21,22

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[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

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"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
25 July 2003	06/08/2003

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL – 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Lehnert, A

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	KIDO J ET AL: "MULTILAYER WHITE LIGHT-EMITTING ORGANIC ELECTROLUMINESCENT DEVICE" SCIENCE, AMERICAN ASSOCIATION FOR THE ADVANCEMENT OF SCIENCE,, US, vol. 267, no. 5202, 3 March 1995 (1995-03-03), pages 1332-1334, XP000857831 ISSN: 0036-8075 the whole document ---	1-4
X	DATABASE WPI Section EI, Week 200065 Derwent Publications Ltd., London, GB; Class X26, AN 2000-669018 XP002249165 -& JP 2000 277259 A (MATSUSHITA DENKI SANGYO KK), 6 October 2000 (2000-10-06) abstract ---	1-4
X	EP 1 074 600 A (SUMITOMO CHEMICAL CO) 7 February 2001 (2001-02-07) page 21, line 19 - line 21 ---	1-4
Y	WO 00 32717 A (KATHIRGAMANATHAN POOPATHY ;SOUTH BANK UNIV ENTPR LTD (GB)) 8 June 2000 (2000-06-08) cited in the application examples 1,8 ---	5,6
Y	PATENT ABSTRACTS OF JAPAN vol. 2000, no. 06, 22 September 2000 (2000-09-22) -& JP 2000 091078 A (MINOLTA CO LTD), 31 March 2000 (2000-03-31) abstract ---	7
Y	GAO XI-CUN ET AL: "Photoluminescence and electroluminescence of a series of terbium complexes" SYNTHETIC METALS, ELSEVIER SEQUOIA, LAUSANNE, CH, vol. 99, no. 2, 12 February 1999 (1999-02-12), pages 127-132, XP002216364 ISSN: 0379-6779 the whole document ---	8,9,14
Y	WO 02 20692 A (ETCHELLS MARK ;CHRISTOU VICTOR (GB); ISIS INNOVATION (GB); MOSLEY) 14 March 2002 (2002-03-14) claims 1,19 ---	8,9

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	GAO D-Q ET AL: "An organic electroluminescent device made from a gadolinium complex" SOLID STATE COMMUNICATIONS, OXFORD, GB, vol. 121, no. 2-3, 2002, pages 145-147, XP002216365 ISSN: 0038-1098 the whole document	8,9
Y	ZHU W ET AL: "Red electroluminescence from a novel europium beta -diketone complex with acylpyrazolone ligand" SYNTHETIC METALS, ELSEVIER SEQUOIA, LAUSANNE, CH, vol. 111-112, 15 May 1999 (1999-05-15), pages 445-447, XP002216366 ISSN: 0379-6779 the whole document	8,9
Y	WO 00 44851 A (KATHIRGAMANATHAN POOPATHY ;SOUTH BANK UNIV ENTPR LTD (GB)) 3 August 2000 (2000-08-03) cited in the application examples 1,2	11,12

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No
PCT/GB 03/01932

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